

Shift charge and spin photocurrents in Dirac surface states of topological insulator

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(Dated: July 14, 2016)

The generation of photocurrent in condensed matter is of main interest for photovoltaic and optoelectronic applications. Shift current, a nonlinear photoresponse, has attracted recent intensive attention as a dominant player of bulk photovoltaic effect in ferroelectric materials. In three dimensional topological insulators Bi_2X_3 (X: Te, Se), we find that Dirac surface states with a hexagonal warping term carry shift current by linearly polarized light. In addition, shift spin-current is introduced with the time-reversal symmetry breaking perturbation. The estimate for the magnitudes of the shift charge- and spin-currents are $0.13I_0$ and $0.21I_0$ (nA/m) with the intensity of light I_0 measured in (W/m^2), respectively, which can offer a useful method to generate these currents efficiently.

Introduction and background- Interaction of light with topological matters has drawn keen attention in condensed matter community. Not only the characterization of topological matter has been done by light-assisted experimental tools that probe electronic bands [1–3] and its spin textures [4, 5], optical conductivities, Kerr rotation [6–10], Farady effect [8–11], etc, but also light plays active roles to dynamically induce topological phase transitions [12–17] and generates a new type of topological excitations [18].

Among them, photocurrent generation in condensed matters is an active research field closely related to light-harvesting energy research [19–23] and optoelectronic device applications [24–29]. Particularly, shift current is the nonlinear optical responses, where the d.c. current is produced by light irradiation without the external electric field or potential gradient, whose direction is determined by the crystal structure or the direction of the electric polarization. It is suggested to play a dominant role in bulk photovoltaic effect of ferroelectric materials including hybrid perovskite materials [20, 21].

Further optimal engineering of materials to carry shift current is under studies [29] to enhance photovoltaic efficiency. The shift current is allowed in noncentrosymmetric systems since it is proportional to the square of the electric field \mathbf{E} . Specifically, it is of topological origin that is driven by the Berry phase connection $\mathbf{a}_n(\mathbf{k})$ of Bloch wavefunctions (n : band index, \mathbf{k} : crystal momentum) and is allowed in noncentrosymmetric systems. This is because the Berry curvature $\mathbf{b}_n(\mathbf{k}) = \nabla_{\mathbf{k}} \times \mathbf{a}_n(\mathbf{k})$ satisfies $\mathbf{b}_n(\mathbf{k}) = -\mathbf{b}_n(-\mathbf{k})$ due to the time-reversal symmetry \mathcal{T} . The inversion symmetry \mathcal{I} further gives the constraint $\mathbf{b}_n(\mathbf{k}) = -\mathbf{b}_n(\mathbf{k})$, which concludes $\mathbf{b}_n(\mathbf{k}) = \mathbf{0}$ and one can choose the gauge $\mathbf{a}_n(\mathbf{k}) = \mathbf{0}$. Therefore, it is essential to break either \mathcal{T} or \mathcal{I} for the nonzero $\mathbf{a}_n(\mathbf{k})$. The physical meaning of the Berry connection $\mathbf{a}_n(\mathbf{k})$ is the intracell coordinates of the electron in the band n , which are related to the electric polarization \mathbf{P} in

ferroelectrics [30, 31]. Therefore, the difference between $\mathbf{a}_{n=c}(\mathbf{k})$ and $\mathbf{a}_{n=v}(\mathbf{k})$ represents the shift of the electrons associated with the optical transitions from the valence band v to the conduction band c . Under steady optical pumping, the constantly induced polarization described above results in the dc current, which is the shift current.

Topological surface states are good candidates to support shift currents as the inversion symmetry \mathcal{I} is always broken at the surface of materials. We show in this paper that linearly polarized light can generate shift currents on warped topological surface states [32, 33] in Bi_2X_3 -type topological insulators. Here the warping term breaks the mirror symmetry in one direction, and this determines unique combination of current and electric field directions to have nonzero shift currents. Furthermore, we propose in this paper the shift spin-current which is also of topological origin described by Berry phase connection. As for the surface states of topological insulators, we propose that pure shift spin-current (without shift charge-current) is induced by breaking the time-reversal symmetry \mathcal{T} with magnetic doping. Its spin direction can be controlled by the direction of surface magnetization in topological insulators. Equipped with a shift charge- and spin-current production on Dirac surface states, improved platforms for opto-electronic and opto-spintronic devices can be provided by topological insulators.

Model and formalism for shift charge- and spin-currents- This nonlinear direct photocurrent can be understood in several ways as explained below. To discuss in concrete terms, let us consider a two-band system under a periodic time-dependent electric field with frequency Ω , $\vec{E}(t) = -\partial_t \vec{A}(t) = -i\Omega(\vec{A}e^{i\Omega t} + \text{c.c.})$. Treating the external field classically, we can write a Floquet Hamiltonian in the basis of valence and conduction bands [34] (hereafter, $\hbar = 1$ and $e = 1$ is used):

$$H_F = \begin{pmatrix} \epsilon_v + \hbar\Omega & i\vec{A}^* \cdot \vec{v}_{vc} \\ -i\vec{A} \cdot \vec{v}_{cv} & \epsilon_c \end{pmatrix}, \quad (1)$$

where we concentrate on one valence band with a single photon absorbed and one conduction band. $\epsilon_{c,v}$ are eigenvalues of original Hamiltonian H . Two bands in the Floquet Hamiltonian are coupled by time-dependent terms, $\langle u_c | \frac{1}{T} \int_0^T H(\vec{k} - \vec{A}(t)) e^{i\Omega t} dt | u_v \rangle$. By taking the linear order of external field only, the coupling can be expressed in terms of Fermi velocity: $-i\vec{A} \cdot \vec{v}_{cv} = -i \sum_j A_j v_{cv}^j$ with $v_{cv}^j = \langle \psi_c | \partial_{k_j} H | \psi_v \rangle = |v_{cv}^j| e^{i\varphi_{cv}^j}$. Sipe and Shkrebtii [35] obtained a simplified expression for a linearly polarized light case by computing photocurrent perturbatively, $J_j^{shift} = \sum_{i=x,y} \chi_j^{ii} E_i E_i$:

$$\chi_j^{ii} = \frac{\pi}{\Omega^2} \int \frac{d^d \vec{k}}{(2\pi)^d} \delta(\omega_{cv} - \Omega) |v_{vc}^i|^2 (\partial_{k_j} \varphi_{cv}^i + a_c^j - a_v^j) \quad (2)$$

where i and j are cartesian coordinates, and Berry connection $a_c^j = -i \langle \psi_c | \partial_{k_j} \psi_c \rangle$ and $a_v^j = -i \langle \psi_v | \partial_{k_j} \psi_v \rangle$ for conduction and valence band, respectively. The contributions are from the states satisfying the resonant condition: $\omega_{cv} - \Omega = \epsilon_c - \epsilon_v - \Omega$. The bracket on the right side is called a shift vector as the Berry connection a_c^j indicates a shifting in real space of conduction band Bloch wave function along j_{th} direction. $(a_c^j - a_v^j)$ is a difference of shifting between conduction and valence band, and $\partial_{k_j} \varphi_{cv}^i$ maintains the gauge invariance. As a result, a shift current is originated from the Bloch wave function's position change in real space due to electronic excitation from a valence band to a conduction band.

An alternative derivation of the photocurrent expression was obtained by two of the authors previously [34, 36] by using Floquet formalism with Keldysh Green's function. They re-derived the three second-order photocurrents in easier manner compared to previous approaches [35, 37, 38]. In particular, the expression of shift current for a general gauge vector \vec{A} is:

$$J_j = \int \frac{d^d k}{(2\pi)^d} \Im \left[\delta(\omega_{cv} - \Omega) (\vec{A}^* \cdot \vec{v}_{vc}) (\vec{A} \cdot \partial_{k_j} \vec{v}) \right] \quad (3)$$

where

$$(\vec{A} \cdot \partial_{k_j} \vec{v})_{cv} = \sum_i A_i \langle u_c | \partial_{k_j} v^i | u_v \rangle \quad (4)$$

Provided that [34] $(\partial_{k_j} v^i)_{cv} = \partial_{k_j} v_{cv} - \langle \partial_{k_j} u_c | v | u_v \rangle - \langle u_c | v | \partial_{k_j} u_v \rangle$, one can recover the shift current expression found by Sipe and Shkrebtii [35] in terms of Berry connections. Expressed in terms of v_{vc}^i and $(\partial_{k_j} v^i)_{cv}$, Eq.(3) just looks like a correlation function between paramagnetic current and diamagnetic current. A Kubo conductivity in the linear response theory is obtained by taking a correlation function between two paramagnetic currents, $j_{para}^i = \frac{\partial H(\vec{k} - \vec{A}(t))}{\partial A_i} \Big|_{\vec{A}=0} = -v^i$. Eq.(3) is still a current-current correlation function, however, since a diamagnetic current contains a gauge vector,

$j_{j,dia}^l = \frac{\partial^2 H(\vec{k} - \vec{A}(t))}{\partial A_l \partial A_j} \Big|_{\vec{A}=0} A_l = (\partial_{k_j} v^l) A_l$, the photocurrent J is proportional to the square of electric field. Note that the three point correlation function of paramagnetic current also gives the second order photocurrent response as injection currents are obtained by Hosur [39]. In contrast, the susceptibility of current $J_j = \sum_{il} \chi_j^{il} E_i E_l$ in Eq.(3) can be computed by the following one-loop integration:

$$\chi_j^{il} = \frac{1}{\Omega^2} \int dk Tr [v^i G(\omega + \Omega, \vec{k}) (\partial_{k_j} v^l) G(\omega, \vec{k})], \quad (5)$$

where $\int dk = \int d^d \vec{k} d\omega / (2\pi)^d$ and $G(\omega, \vec{k}) = [\omega - H]^{-1}$. We want to emphasize that the diamagnetic current, $\partial_{k_j} \hat{j}^l$, is responsible for the shifting of charge center, and it contains the index of photocurrent direction.

Lastly, we introduce a shift spin-current, which is an extension of a shift charge-current discussed so far. Having considered the shifting of charge center upon excitation from a valence to a conduction band, one can also think of the shifting of spin center that leads a shift spin-current. This can be most conveniently obtained from the one-loop diagram formulation: we take a particular spin direction, \hat{s}_n , for a diamagnetic current response, while we keep a paramagnetic current to be coupled to external electric field:

$$\chi_{j,\hat{s}_n}^{il} = \frac{1}{\Omega^2} \int dk Tr [v^i G(\omega + \Omega, \vec{k}) \{\hat{s}_n, (\partial_{k_j} v^l)\} G(\omega, \vec{k})], \quad (6)$$

where if $\hat{s}_n = \sigma_0/2$, this will recover the previous expression, Eq.(5). Young and Rappe [40] first discussed the spin bulk-photovoltaic effect in antiferromagnets by computing shift current for spin-up and spin-down, independently. Our expression, Eq.(6), is applicable to more general cases with spin-orbit couplings.

In the following, we explicitly show that the breaking of the inversion symmetry \mathcal{I} is required to have a shift charge-current, and the breaking of the time-reversal symmetry \mathcal{T} is additionally required to have a shift spin-current. To demonstrate, we consider two examples in 3d topological insulators: warped Dirac surface states carry shift charge-currents, and massive Dirac surface states with band-bending carry shift spin-currents.

Shift charge current in Dirac surface states of topological insulators- A system with the inversion symmetry carries a zero shift current. We first review this fact. This is intuitively correct, because a state $|k\rangle$ and its inversion symmetric partner $\mathcal{P}|k\rangle$ will be shifted in opposite direction after being excited to a conduction band. More formally, the inversion symmetry gives the following relation:

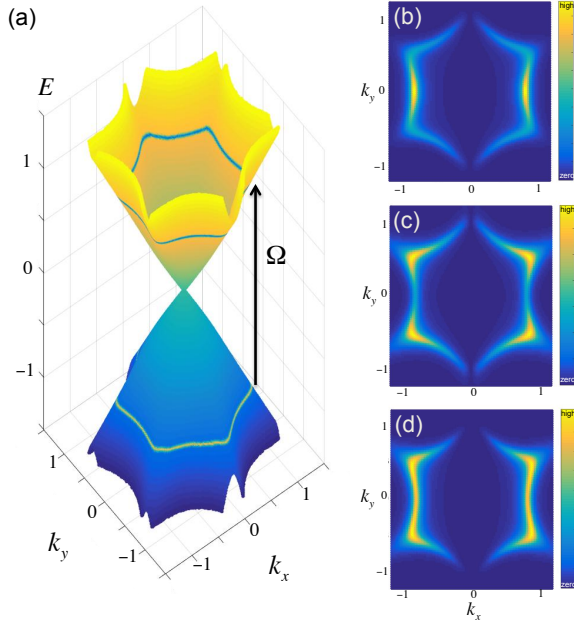


FIG. 1. The susceptibility of shift charge-current, $J_{y, \text{warp}} = \chi_{yy}^{yy} E_y^2$, is decomposed into the parts of perturbations and responses. (a) Warped Dirac surface states with resonance condition in distinct colors. $k_{x,y}$ and E are in unit of $\sqrt{v_F/\lambda}$ and $\sqrt{v_F^3/\lambda}$, respectively. (b) The coupling strength of $|\vec{k}|$ to a diamagnetic current response, $\delta(\omega_{cv} - \Omega)|(\partial_{k_y} v^y)_{cv}|$, is plotted. (c) The coupling strength of $|\vec{k}|$ to an external electric field, $\delta(\omega_{cv} - \Omega)|(v^y)_{vc}|$, is plotted. (d) The contribution of shift charge-current from $|\vec{k}|$, $\delta(\omega_{cv} - \Omega)|(v^y)_{vc}(\partial_{k_y} v^y)_{cv}|$, is plotted.

$$\begin{aligned}
 v_{vc}^i(\vec{k}) \partial_{k_j} v_{cv}^l(\vec{k}) &= (\partial_{k_i} H(\vec{k}))_{vc} (\partial_{k_j} \partial_{k_l} H(\vec{k}))_{cv}, \\
 &\xrightarrow{\mathcal{I}} (-\partial_{k_i} H(-\vec{k}))_{vc} (\partial_{k_j} \partial_{k_l} H(-\vec{k}))_{cv}, \\
 &= -v_{vc}^i(-\vec{k}) (\partial_{k_j} v^l(-\vec{k}))_{cv}. \quad (7)
 \end{aligned}$$

In the second equality, we use the inversion symmetry relation of Hamiltonian: $H(\vec{k}) = e^{-ikr} \hat{\mathcal{H}} e^{ikr} \xrightarrow{\mathcal{I}} e^{-ikr} \mathcal{P}^{-1} \hat{\mathcal{H}} \mathcal{P} e^{ikr} = \mathcal{P}^{-1} H(-\vec{k}) \mathcal{P}$ (see supplementary information). Therefore,

$$\begin{aligned}
 \chi_j^{il} &= \frac{1}{2} \int \frac{d^d \vec{k}}{(2\pi)^d} \delta(\omega_{cv} - \Omega) \Im \left[v_{vc}^i(\vec{k}) (\partial_{k_j} v^l(\vec{k}))_{cv} \right. \\
 &\quad \left. + v_{vc}^i(-\vec{k}) (\partial_{k_j} v^l(-\vec{k}))_{cv} \right] = 0 \quad (8)
 \end{aligned}$$

Note that, since the spin is invariant under the inversion symmetry operation, $\mathcal{I} : \hat{s}_n \rightarrow \hat{s}_n$, the susceptibility of shift spin-current is also exactly cancelled between inversion symmetry partners.

Now we study Dirac surface states in 3d topological insulators that are localized near open surfaces. Its low energy effective Hamiltonian is $H_0 = -v_F k_y \sigma_x + v_F k_x \sigma_y$,

for which the inversion symmetry is absent. But, $H_0(\vec{k} - \vec{A}(t))$ does not contain diamagnetic terms ($\sim A_i A_i$), hence no shift current. With warping effects induced by C_{3v} crystalline symmetry in discovered 3d topological insulators of Bi_2X_3 (where $\text{X} = \text{Te}, \text{Se}$) type [32], we find that shift charge-currents are non-zero. The warped Dirac surface state Hamiltonian is [33]:

$$H = -v_F k_y \sigma_x + v_F k_x \sigma_y + \lambda(k_x^3 - 3k_x k_y^2) \sigma_z. \quad (9)$$

This Hamiltonian is mirror symmetric along x , but not along y . As a result, the susceptibility χ_j^{il} ($j, i, l = x, y$) can be non-zero when the number of index y appears odd times so that that its inversion symmetry partner does not cancel the shift current. Indeed, a straightforward calculation of current-current correlation function Eq.(5) shows that non-zero components are (see supplementary information):

$$\chi_y^{xx} = \chi_x^{yx} = -\chi_y^{yy} = \frac{3\lambda}{16v_F^2} \quad (10)$$

Upon a linearly polarized light $\vec{E}(t) = E_0(\cos \phi \hat{x} + \sin \phi \hat{y}) \cos \Omega t$, an induced shift current is:

$$\begin{aligned}
 \vec{J}_{\text{warp}} &= -\frac{3\lambda}{16v_F^2} [E_y^2 \hat{y} - E_x^2 \hat{x} - E_x E_y \hat{x} - E_y E_x \hat{y}] \\
 &= \frac{3\lambda E_0^2}{16v_F^2} [\sin 2\phi \hat{x} + \cos 2\phi \hat{y}] \quad (11)
 \end{aligned}$$

Interestingly, the shift charge-photocurrent found here is photon-frequency independent. This is in a sharp contrast to the injection photocurrent calculation [41] based on the Fermi's golden rule, where a photocurrent by linearly polarized light is a function of frequency Ω .

Our numerical estimation is $J_{\text{warp}} = 0.13 I_0 (nA/m)$, where I_0 is the intensity of light (see supplementary information), using warping strength and Fermi velocity estimated by Fu [33]. The photocurrent value is comparable to the theoretical estimation of circular photogalvanic current (CPGC) by Hosur [39] where photocurrent $0.1 I_0 (nA/m)$ is estimated for $10(T)$ in-plane magnetic field, which is to break the in-plane rotational symmetry. Note that the generation of CPGC is based on the selective excitation from a valence band to a conduction band, and this can be alternatively computed using the Fermi's golden rule [41].

In experiments [42–44], photocurrents are measured in Dirac surface states using oblique incident light, which is again to break the in-plane rotational symmetry. Assuming the sample size as $1(mm^2)$, the photocurrent density is in the order of $1 I_0 (pA/m)$, which is two orders of magnitude lower than the theoretical estimation of shift currents on warped Dirac surface states.

Shift spin-current in Dirac surface states of topological insulator with magnetic ordering- A shift spin-current

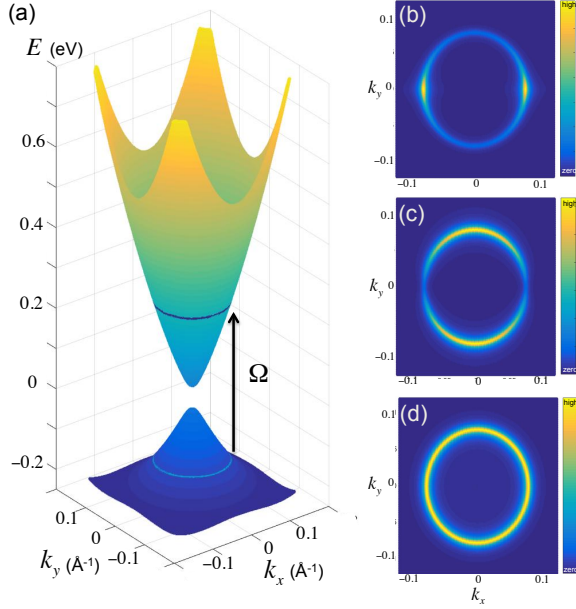


FIG. 2. The susceptibility of shift spin-current, $J_{x,spin} = \chi_{x,\hat{s}_x}^{xx} E_x^2$, is decomposed into the parts of perturbations and responses. (a) The dispersion of gapped Dirac surface states with resonance condition in distinct colors. $m = 0.03(\text{eV})$, $g = 7(\text{eV}\text{\AA}^2)/\hbar^2$, $v_F = 2.55(\text{eV}\text{\AA})/\hbar$ are used. (b) The coupling strength of $|\vec{k}\rangle$ to a spin-diamagnetic current response, $\delta(\omega_{cv} - \Omega)|(\{\hat{s}_x, \partial_{k_x} v^x\})_{cv}|$, is plotted. (c) The coupling strength of $|\vec{k}\rangle$ to an external electric field, $\delta(\omega_{cv} - \Omega)|(\{v^x\}_{vc})|$, is plotted. (d) The contribution of shift spin-current from $|\vec{k}\rangle$, $\delta(\omega_{cv} - \Omega)|(\{v^x\}_{vc}(\{\hat{s}_x, \partial_{k_x} v^x\})_{cv})|$, is plotted.

generated by one state is exactly cancelled by its time-reversal partner state. Thus, the absence of both inversion and time-reversal symmetries is required for a system to carry shift spin-currents. Specifically, due to the time-reversal operation,

$$\begin{aligned} v^i(\vec{k})_{vc}(\partial_{k_j} v^l(\vec{k}))_{cv} &= (\partial_{k_i} H(\vec{k}))_{vc}(\partial_{k_j} \partial_{k_l} H(\vec{k}))_{cv}, \\ &\xrightarrow{\mathcal{T}} (-\partial_{k_i} H(-\vec{k}))_{vc}^* (\partial_{k_j} \partial_{k_l} H(-\vec{k}))_{cv}^*, \\ &= -v^i(-\vec{k})_{vc}^* (\partial_{k_j} v^l(-\vec{k}))_{cv}^*. \end{aligned} \quad (12)$$

where the time-reversal operation is meant to be $H(\vec{k}) = e^{-ikr} \hat{\mathcal{H}} e^{ikr} \xrightarrow{\mathcal{T}} e^{-ikr} \Theta^{-1} \hat{\mathcal{H}} \Theta e^{ikr} = \sigma^y H^*(-\vec{k}) \sigma^y$ (see supplementary information). Combined with the spin operator, which is to pick a diamagnetic current along a particular spin direction:

$$v_{vc}^i(\vec{k})(\{\hat{s}_n, \partial_{k_j} v^l(\vec{k})\})_{cv} \xrightarrow{\mathcal{T}} (v^i(-\vec{k}))_{vc}^* (\{\hat{s}_n, \partial_{k_j} v^l(-\vec{k})\})_{cv}^*.$$

As before, the summation over \vec{k} can be done for $|\vec{k}\rangle$ and $|\vec{k}\rangle$. Taking the imaginary part of the integrand leads to the zero shift spin-susceptibility:

$$\begin{aligned} \chi_{j,spin}^{il} &= \frac{1}{2} \int \frac{d^d \vec{k}}{(2\pi)^d} \delta(\omega_{cv} - \Omega) \Im \left[v_{vc}^i(\vec{k})(\{\hat{s}_n, \partial_{k_j} v^l(\vec{k})\})_{cv} \right. \\ &\quad \left. + v_{vc}^i(-\vec{k})(\{\hat{s}_n, \partial_{k_j} v^l(-\vec{k})\})_{cv} \right] = 0. \end{aligned} \quad (13)$$

This is the case for a linearly polarized light considered in this study, which preserves the time-reversal symmetry. A simple example of a Dirac surface state with perturbations that generate a shift spin-current with zero charge-current is following:

$$H = -v_F k_y \sigma_x + v_F k_x \sigma_y + m \sigma_z + g(k_x^2 + k_y^2) \sigma_0. \quad (14)$$

The third term on the right side is from the magnetic ordering on the surface of 3d topological insulator that breaks the time-reversal symmetry. The last term is a band-bending term that provides diamagnetic terms ($\sim A_i A_l$). Computing the susceptibilities for different combinations of indices, we obtain the following non-zero components:

$$-\chi_{x,\hat{s}_x}^{xx} = \chi_{x,\hat{s}_y}^{yx} = -\chi_{y,\hat{s}_x}^{xy} = \chi_{y,\hat{s}_y}^{yy} = \frac{gm}{2v_F \Omega^2} \quad (15)$$

for the frequency of photon larger than the energy gap, $\Omega > m$. As opposed to the warped Dirac surface state example where we only keep the leading order of warping strength λ , Eq.(15) is exact results for Hamiltonian Eq.(14). Note that the spin direction of photocurrent rotate with the direction of magnetic ordering, $m\sigma_z$. Together, a linearly polarized light, $\vec{E}(t) = E_0(\cos \phi \hat{x} + \sin \phi \hat{y}) \cos \Omega t$, induces the following shift spin-current:

$$\vec{J}_{spin} = -\frac{gmE_0^2}{2v_F \Omega^2} (\cos \phi \hat{x} + \sin \phi \hat{y})(\cos \phi \hat{s}_x + \sin \phi \hat{s}_y) \quad (16)$$

The band-bending near the surface of topological insulators have been reported in several studies [5, 45, 46]. By introducing a magnetic dopants providing Zeeman term, Hamiltonian (14) is readily realized. In contrast to the shift charge-current in warped Dirac surface states, we have more flexibility to control the direction of magnetic ordering and its magnitude. With reasonable choices of $m = 30(\text{meV})$, $\hbar\Omega = 60(\text{meV})$ and experimentally observed Fermi velocity and band bending parameters [46], we obtain shift spin-current density $0.21 I_0 (\text{nA}/m)$, which is in the same order with the shift charge-current estimated in the previous section.

On the other hand, a large band-bending at the surface of topological insulators induces a 2d electron gas (2DEG) with Rashba spin splitting. The 2DEG is described by the same Hamiltonian (14) with different

Fermi velocity $v_F = 0.36\hbar^{-1}(eV\text{\AA})$, one order of magnitude less than that for Dirac surface state (see supplementary information). As a result, we obtain the 2DEG spin-shift current density $1.5I_0(nA/m)$, which is even larger than that of massive Dirac surface states. By carefully tuning the Fermi energy, one will be able to see different involvement of Dirac surface states and 2DEG in the production of shift spin-currents.

In summary, starting from the second order photocurrent derivation in the Floquet formalism, shift charge- and spin-currents are explained as two point correlation functions of para- and dia-magnetic currents. Two examples, both from the Dirac surface states, are shown and their theoretical values in experiments are quantified.

This work was supported by the EPIQS initiative of the Gordon and Betty Moore Foundation (TM) and by JSPS Grant-in-Aid for Scientific Research (No. 24224009, and No. 26103006) from MEXT, Japan, and ImPACT Program of Council for Science, Technology and Innovation (Cabinet office, Government of Japan) (NN).

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Supplementary Materials

S1. Shift currents and symmetries

In the main text, we claim that a shift charge-current is allowed in the absence of the inversion symmetry, and a shift spin-current requires, in addition, the absence of the time-reversal symmetry. In this supplementary, we provide the details of proofs.

Inversion symmetry and shift charge-currents

First of all, consider a system with the inversion symmetry. The inversion symmetric Hamiltonian satisfies $[\hat{\mathcal{H}}, \mathcal{P}] = 0$, which connects Bloch Hamiltonian $H(\vec{k})$ and $H(-\vec{k})$:

$$H(\vec{k}) = e^{-i\vec{k}\cdot\vec{r}} \hat{\mathcal{H}} e^{i\vec{k}\cdot\vec{r}} = e^{-i\vec{k}\cdot\vec{r}} \mathcal{P}^{-1} \hat{\mathcal{H}} \mathcal{P} e^{i\vec{k}\cdot\vec{r}} \quad (17)$$

$$= \mathcal{P}^{-1} e^{i\vec{k}\cdot\vec{r}} \hat{\mathcal{H}} e^{-i\vec{k}\cdot\vec{r}} \mathcal{P} = \mathcal{P}^{-1} H(-\vec{k}) \mathcal{P} \quad (18)$$

According to this, we can establish the relation of Fermi velocity between inversion symmetry partners:

$$v_{vc}^i(\vec{k}) = \langle u_v(\vec{k}) | \frac{\partial H(\vec{k})}{\partial k_i} | u_c(\vec{k}) \rangle = \langle u_v(\vec{k}) | \frac{H(\vec{k} + \Delta k \hat{i}) - H(\vec{k})}{\Delta k} | u_c(\vec{k}) \rangle \quad (19)$$

$$= -\langle u_v(\vec{k}) | \mathcal{P}^{-1} \frac{H(-\vec{k} - \Delta k \hat{i}) - H(-\vec{k})}{-\Delta k} \mathcal{P} | u_c(\vec{k}) \rangle = -\langle u_v(\vec{k}) | \mathcal{P}^{-1} \frac{\partial H(-\vec{k})}{\partial k_i} \mathcal{P} | u_c(\vec{k}) \rangle \quad (20)$$

$$= -v_{vc}(-\vec{k}) \quad (21)$$

where in the last equality, we used the fact that $\mathcal{P} | u_{c,v}(\vec{k}) \rangle$ is an eigenstate of $H(-\vec{k})$. Similarly, it is straightforward to show $(\partial_{k_j} v^l(\vec{k}))_{cv} = (\partial_{k_j} v^l(-\vec{k}))_{cv}$:

$$(\partial_{k_j} v^l(\vec{k}))_{cv} = \langle u_c(\vec{k}) | \frac{\partial^2 H(\vec{k})}{\partial k_j \partial k_l} | u_v(\vec{k}) \rangle \quad (22)$$

$$= \langle u_c(\vec{k}) | \mathcal{P}^{-1} \frac{\partial^2 H(-\vec{k})}{\partial k_j \partial k_l} \mathcal{P} | u_v(\vec{k}) \rangle = (\partial_{k_j} v^l(-\vec{k}))_{cv} \quad (23)$$

Therefore, the contribution to the shift charge-current from a Bloch state $|\vec{k}\rangle$ is exactly cancelled by its inversion symmetry partner $\mathcal{P}|\vec{k}\rangle$:

$$v_{vc}(\vec{k})(\partial_{k_j} v^l(\vec{k}))_{cv} = -v_{vc}(-\vec{k})(\partial_{k_j} v^l(-\vec{k}))_{cv} \quad (24)$$

Therefore,

$$\chi_j^{il} = \frac{1}{2} \int d\vec{k} \delta(d_z) \Im \left[v_{vc}^i(\vec{k}) \partial_{k_j} v_{cv}^l(\vec{k}) + v_{vc}^i(-\vec{k}) \partial_{-k_j} v_{cv}^l(-\vec{k}) \right] = 0$$

Let us comment that the inversion symmetry operation on shift spin-current. Because $\mathcal{I} : \hat{s}_n \rightarrow \hat{s}_n$,

$$(\{\hat{s}_n, \partial_{k_j} v^l(\vec{k})\})_{vc} = \langle u_v(\vec{k}) | \left\{ \hat{s}_n, \frac{\partial^2 H(\vec{k})}{\partial k_j \partial k_l} \right\} | u_c(\vec{k}) \rangle \quad (25)$$

$$= \langle u_v(\vec{k}) | \mathcal{P}^{-1} \left\{ \hat{s}_n, \frac{\partial^2 H(-\vec{k})}{\partial k_j \partial k_l} \right\} \mathcal{P} | u_c(\vec{k}) \rangle \quad (26)$$

$$= (\{\hat{s}_n, \partial_{k_j} v^l(-\vec{k})\})_{vc} \quad (27)$$

Therefore,

$$v(\vec{k})_{vc}(\{\hat{s}_n, \partial_{k_j} v^l(\vec{k})\})_{cv} = -v(-\vec{k})_{vc}(\{\hat{s}_n, \partial_{k_j} v^l(-\vec{k})\})_{cv} \quad (28)$$

Thus, the presence of inversion symmetry gives the exactly opposite shift spin-current contributions from its inversion symmetry partners, and the inversion symmetry must be broken for a system to carry a shift spin-current as well.

The time-reversal symmetry and shift currents

Then, let us consider a system with the time-reversal symmetry, $[\Theta, \hat{\mathcal{H}}] = 0$. The time reversal operation can be written $\Theta = i\sigma_y K$, where operator K is complex conjugation. The Bloch wavefunction is transformed in the following way:

$$H(\vec{k}) = e^{-i\vec{k} \cdot \vec{r}} \hat{\mathcal{H}} e^{i\vec{k} \cdot \vec{r}} = e^{-i\vec{k} \cdot \vec{r}} \Theta^{-1} \hat{\mathcal{H}} \Theta e^{i\vec{k} \cdot \vec{r}} \quad (29)$$

$$= i\sigma_y K e^{i\vec{k} \cdot \vec{r}} \hat{\mathcal{H}} e^{-i\vec{k} \cdot \vec{r}} (-i) \sigma_y K \quad (30)$$

$$= \sigma_y e^{-i\vec{k} \cdot \vec{r}} \hat{\mathcal{H}}^* e^{i\vec{k} \cdot \vec{r}} \sigma_y \quad (31)$$

$$= \sigma_y \left(e^{i\vec{k} \cdot \vec{r}} \hat{\mathcal{H}} e^{-i\vec{k} \cdot \vec{r}} \right)^* \sigma_y \quad (32)$$

$$= \sigma_y H^*(-\vec{k}) \sigma_y \quad (33)$$

$$(34)$$

Accordingly, Fermi velocity component is transformed:

$$v_{vc}^i(\vec{k}) = \langle u_v(\vec{k}) | \frac{\partial H(\vec{k})}{\partial k_i} | u_c(\vec{k}) \rangle = \langle u_v(\vec{k}) | \frac{H(\vec{k} + \Delta k \hat{i}) - H(\vec{k})}{\Delta k} | u_c(\vec{k}) \rangle \quad (35)$$

$$= -\langle u_v(\vec{k}) | \sigma_y \frac{H^*(-\vec{k} - \Delta k \hat{i}) - H^*(-\vec{k})}{-\Delta k} \sigma_y | u_c(\vec{k}) \rangle \quad (36)$$

$$= -\langle u_v(\vec{k}) | \sigma_y \frac{\partial H^*(-\vec{k})}{\partial k_i} \sigma_y | u_c(\vec{k}) \rangle \quad (37)$$

$$= -\left({}^* \langle u_v(\vec{k}) | \sigma_y \frac{\partial H(-\vec{k})}{\partial k_i} \sigma_y | u_c(\vec{k}) \rangle {}^* \right) \quad (38)$$

$$= -\left(\langle u_v(\vec{k}) | \Theta^{-1} \frac{\partial H(-\vec{k})}{\partial k_i} \Theta | u_c(\vec{k}) \rangle \right) \quad (39)$$

$$= -v_{vc}^*(-\vec{k}) \quad (40)$$

where we use the fact that the time reversal partner of a state $|\vec{k}\rangle$ is an eigen state at $-\vec{k}$. Similarly, one can prove that

$$(\partial_{k_j} v^l(\vec{k}))_{cv} = \langle u_c(\vec{k}) | \frac{\partial^2 H(\vec{k})}{\partial k_j \partial k_l} | u_v(\vec{k}) \rangle \quad (41)$$

$$= \langle u_c(\vec{k}) | \sigma_y \frac{\partial^2 H^*(-\vec{k})}{\partial k_j \partial k_l} \sigma_y | u_v(\vec{k}) \rangle = (\partial_{k_j} v^l(-\vec{k}))_{cv}^* \quad (42)$$

This proves the claim in the main body.

S2. Spherical coordinate expressions

Spherical coordinate is convenient to deal with resonant conditions in Dirac Hamiltonians. It converts the momentum integration to spherical angular integration with a fixed radius. Consider a Hamiltonian:

$$H = (h_1, h_2, h_3) \cdot \vec{\sigma}, \quad (43)$$

$$= h(\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta) \cdot \vec{\sigma}. \quad (44)$$

Its eigenvectors are:

$$|u_c\rangle = \begin{pmatrix} e^{-i\phi} \cos \theta/2 \\ \sin \theta/2 \end{pmatrix}, \quad |u_v\rangle = \begin{pmatrix} e^{-i\phi} \sin \theta/2 \\ -\cos \theta/2 \end{pmatrix}. \quad (45)$$

In this gauge choice, let us compute off-diagonal components of pauli matrices:

$$\langle u_c | \sigma_1 | u_v \rangle = -\cos \phi \cos \theta - i \sin \phi \quad (46)$$

$$\langle u_c | \sigma_2 | u_v \rangle = -\sin \phi \cos \theta + i \cos \phi \quad (47)$$

$$\langle u_c | \sigma_3 | u_v \rangle = \sin \theta \quad (48)$$

These quantities are gauge dependent, but the expression of shift current is gauge-independent. A Jacobian appears as the momentum integration is converted into the spherical coordinate integration,

$$\int dk_x dk_y dh = \int \left| \frac{\partial(k_x, k_y, h_3)}{\partial(h, \theta, \phi)} \right| dh d\theta d\phi \quad (49)$$

In the following examples, the Jacobian is simply $J = h^2 \sin \theta$.

S3. Shift charge-current in warped Dirac surface states

Let us compute shift charge-currents in warped Dirac surface states. First of all, the Hamiltonian and its derivatives are (in this section, $v_F = 1$ is assumed for simplicity):

$$h = [-k_y, k_x, \lambda(k_x^3 - 3k_x k_y^2)] \cdot \vec{\sigma}, \quad (50)$$

$$v^x = \partial_{k_x} h = [0, 1, 3\lambda(k_x^2 - k_y^2)] \cdot \vec{\sigma}, \quad v^y = \partial_{k_y} h = [-1, 0, -6\lambda k_x k_y] \cdot \vec{\sigma}, \quad (51)$$

$$\partial_{k_x} v^x = 6\lambda k_x \sigma_3, \quad \partial_{k_y} v^x = -6\lambda k_y \sigma_3, \quad \partial_{k_x} v^y = -6\lambda k_y \sigma_3, \quad \partial_{k_y} v^y = -6\lambda k_x \sigma_3. \quad (52)$$

Our goal is to compute the susceptibility of shift current, $J_j = \chi_j^{il} E_i E_l$:

$$\chi_j^{il} = \frac{\pi}{\Omega^2} \int \frac{d^2 \vec{k}}{4\pi^2} \delta(d_z) (v^i)_{vc} (\partial_{k_j} v^l)_{cv} \quad (53)$$

Using spherical coordinates:

$$h_1 = h \sin \theta \cos \phi = -k_y \quad (54)$$

$$h_2 = h \sin \theta \sin \phi = k_x \quad (55)$$

$$h_3 = h \cos \theta = \lambda(k_x^3 - 3k_x k_y^2) = \lambda h^3 \sin^3 \theta \cos \phi (\cos^2 \phi - 3 \sin^2 \phi) \quad (56)$$

The 2d momentum integration is converted to spherical coordinate integration with a delta function:

$$\int dk_x dk_y = \int dk_x dk_y dh_3 \delta(h_3 - h \cos \theta) \quad (57)$$

$$= \int \left| \frac{\partial(k_x, k_y, h_3)}{\partial(h, \theta, \phi)} \right| dh d\theta d\phi \frac{1}{h} \delta(\cos \theta - \lambda h^2 \sin^3 \theta \cos \phi (\cos^2 \phi - 3 \sin^2 \phi)) \quad (58)$$

where the Jacobian is $\left| \frac{\partial(k_x, k_y, h_3)}{\partial(h, \theta, \phi)} \right| = h^2 \sin \theta$. Let us compute χ_y^{yy} explicitly:

$$\chi_y^{yy} = \frac{\pi}{\Omega^2} \int \frac{d^2 \vec{k}}{4\pi^2} \delta(\omega_{cv} - \Omega) \Im[(v^y)_{vc} (\partial_{k_y} v^y)_{cv}], \quad (59)$$

$$= \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \delta(2h - \Omega) \frac{1}{h} \delta(\cos \theta + O(\lambda)) \Im[\langle u_v | -\sigma_1 + O(\lambda) | u_c \rangle \langle u_c | -6\lambda k_x \sigma_3 | u_v \rangle] \quad (60)$$

$$\simeq \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \delta(2h - \Omega) \frac{1}{h} \delta(\cos \theta) \Im[\langle u_v | -\sigma_1 | u_c \rangle \langle u_c | 6\lambda h \sin \theta \sin \phi \sigma_3 | u_v \rangle] \quad (61)$$

$$= \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \delta(2h - \Omega) \frac{-6\lambda h \sin \theta \sin \phi}{h} \delta(\cos \theta) \Im[\langle u_v | \sigma_1 | u_c \rangle \langle u_c | \sigma_3 | u_v \rangle] \quad (62)$$

$$= \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \frac{\delta(h - \Omega/2)}{2} \frac{-6\lambda h \sin \theta \sin \phi}{h} \delta(\cos \theta) \Im[(-\cos \phi \cos \theta + i \sin \phi) \sin \theta] \quad (63)$$

$$= -\frac{1}{4\pi\Omega^2} \int d\phi \left(\frac{\Omega}{2} \right)^2 3\lambda \sin^2 \phi \quad (64)$$

$$= -\frac{3\lambda}{16}, \quad (65)$$

where the leading order of λ is kept only. This exactly reproduces the result in the main text with $v_F = 1$. Similarly, other components can be computed in a straightforward manner:

$$\chi_x^{yx} = \frac{\pi}{\Omega^2} \int \frac{d^2 \vec{k}}{4\pi^2} \delta(\omega_{cv} - \Omega) \Im[(v^y)_{vc} (\partial_{k_x} v^x)_{cv}], \quad (66)$$

$$\simeq \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \delta(2h - \Omega) \frac{1}{h} \delta(\cos \theta) \Im[\langle u_v | -\sigma_1 | u_c \rangle \langle u_c | -6\lambda h \sin \theta \sin \phi \sigma_3 | u_v \rangle] = \frac{3\lambda}{16}, \quad (67)$$

$$\chi_x^{xy} = \frac{\pi}{\Omega^2} \int \frac{d^2 \vec{k}}{4\pi^2} \delta(\omega_{cv} - \Omega) \Im[(v^x)_{vc} (\partial_{k_y} v^y)_{cv}], \quad (68)$$

$$\simeq \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \delta(2h - \Omega) \frac{1}{h} \delta(\cos \theta) \Im[\langle u_v | \sigma_2 | u_c \rangle \langle u_c | 6\lambda h \sin \theta \cos \phi \sigma_3 | u_v \rangle] = \frac{3\lambda}{16}, \quad (69)$$

$$\chi_y^{xx} = \frac{\pi}{\Omega^2} \int \frac{d^2 \vec{k}}{4\pi^2} \delta(\omega_{cv} - \Omega) \Im[(v^x)_{vc} (\partial_{k_y} v^x)_{cv}], \quad (70)$$

$$\simeq \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \delta(2h - \Omega) \frac{1}{h} \delta(\cos \theta) \Im[\langle u_v | \sigma_2 | u_c \rangle \langle u_c | 6\lambda h \sin \theta \cos \phi \sigma_3 | u_v \rangle] = \frac{3\lambda}{16}. \quad (71)$$

S4. Shift spin-current in massive Dirac surface states

By introducing magnetic ordering at the surface of 3d topological insulator, the time-reversal symmetry is broken and we find shift spin-currents linearly proportional to band bending parameter. The Hamiltonian and its derivatives are:

$$h = [-k_y, k_x, m] \cdot \vec{\sigma} + g(k_x^2 + k_y^2) \sigma_0, \quad (72)$$

$$v^x = \partial_{k_x} h = \sigma_2 + 2gk_x \sigma_0, \quad v^y = \partial_{k_y} h = -\sigma_x + 2gk_y \sigma_0, \quad (73)$$

$$\partial_{k_x} v^x = 2g\sigma_0, \quad \partial_{k_y} v^y = 2g\sigma_0. \quad (74)$$

For spin shift-current, we need pick a particular spin direction in the diamagnetic current:

$$\{\hat{s}_n, \partial_{k_j} v^l\} = \frac{1}{2} [\sigma_n (\partial_{k_j} v^l) + (\partial_{k_j} v^l) \sigma_n] \quad (75)$$

Our goal is to compute the susceptibility of shift spin-current, $J_{j, \hat{s}_n} = \chi_{j, \hat{s}_n}^{il} E_i E_l$:

$$\chi_{j, \hat{s}_n}^{il} = \frac{\pi}{\Omega^2} \int \frac{d^2 \vec{k}}{(2\pi)^2} \delta(\omega_{cv} - \Omega) \Im[(v^i)_{vc} (\partial_{k_j} \{\hat{s}_n, v^l\})_{cv}], \quad (76)$$

The integration over momentum is converted to spherical coordinate integration with a delta function:

$$\int dk_x dk_y = \int dk_x dk_y dh_3 \delta(h_3 - h \cos \theta) \quad (77)$$

$$= \int \left| \frac{\partial(k_x, k_y, h_3)}{\partial(h, \theta, \phi)} \right| dh d\theta d\phi \frac{1}{h} \delta\left(\cos \theta - \frac{m}{h}\right) \quad (78)$$

where the Jacobian is $\left| \frac{\partial(k_x, k_y, h_3)}{\partial(h, \theta, \phi)} \right| = h^2 \sin \theta$. For example, let us get susceptibility χ_{x, \hat{s}_x}^{xx} :

$$\chi_{x, \hat{s}_x}^{xx} = \frac{\pi}{\Omega^2} \int \frac{d^2 \vec{k}}{4\pi^2} \delta(\omega_{cv} - \Omega) \Im[(v^x)_{vc} (\partial_{k_x} \{\hat{s}_x, v^x\})_{cv}], \quad (79)$$

$$= \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \frac{1}{h} \delta\left(\cos \theta - \frac{h_3}{h}\right) \frac{\delta(h - \Omega/2)}{2} \Im[\langle u_v | \sigma_y + 2k_x s_0 | u_c \rangle \langle u_v | 2g\sigma_x | u_c \rangle] \quad (80)$$

$$= \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h \sin \theta \delta\left(\cos \theta - \frac{h_3}{h}\right) \frac{\delta(h - \Omega/2)}{2} \Im[(-\sin \phi \cos \theta - i \cos \phi) 2g(-\cos \phi \cos \theta - i \sin \phi)] \quad (81)$$

$$= \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h \sin \theta \delta\left(\cos \theta - \frac{h_3}{h}\right) \frac{\delta(h - \Omega/2)}{2} 2g \cos \theta \quad (82)$$

$$= -\frac{gm}{2\Omega^2} \quad (83)$$

Note that there is no approximation taken in this calculation. Therefore, as long as the continuum Hamiltonian is valid, the shift spin-current expression obtained here is exact. Other components can be calculated similarly:

$$\chi_{x, \hat{s}_y}^{yx} = \frac{\pi}{\Omega^2} \int \frac{d^2 \vec{k}}{4\pi^2} \delta(\omega_{cv} - \Omega) \Im[(v^y)_{vc} (\partial_{k_x} \{\hat{s}_y, v^x\})_{cv}], \quad (84)$$

$$= \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \frac{1}{h} \delta\left(\cos \theta - \frac{h_3}{h}\right) \frac{\delta(h - \Omega/2)}{2} \Im[\langle u_v | -\sigma_x | u_c \rangle \langle u_v | 2g\sigma_y | u_c \rangle] = -\frac{gm}{2\Omega^2}, \quad (85)$$

$$\chi_{y, \hat{s}_x}^{xy} = \frac{\pi}{\Omega^2} \int \frac{d^2 \vec{k}}{4\pi^2} \delta(\omega_{cv} - \Omega) \Im[(v^x)_{vc} (\partial_{k_y} \{\hat{s}_x, v^y\})_{cv}], \quad (86)$$

$$= \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \frac{1}{h} \delta\left(\cos \theta - \frac{h_3}{h}\right) \frac{\delta(h - \Omega/2)}{2} \Im[\langle u_v | \sigma_y | u_c \rangle \langle u_v | 2g\sigma_x | u_c \rangle] = \frac{gm}{2\Omega^2}, \quad (87)$$

$$\chi_{y, \hat{s}_y}^{yy} = \frac{\pi}{\Omega^2} \int \frac{d^2 \vec{k}}{4\pi^2} \delta(\omega_{cv} - \Omega) \Im[(v^y)_{vc} (\partial_{k_y} \{\hat{s}_y, v^y\})_{cv}], \quad (88)$$

$$= \frac{1}{4\pi\Omega^2} \int dh d\theta d\phi h^2 \sin \theta \frac{1}{h} \delta\left(\cos \theta - \frac{h_3}{h}\right) \frac{\delta(h - \Omega/2)}{2} \Im[\langle u_v | -\sigma_x | u_c \rangle \langle u_v | 2g\sigma_y | u_c \rangle] = -\frac{gm}{2\Omega^2}, \quad (89)$$

$$(90)$$

Other combinations of indices yield zero susceptibility.

S5. Numerical estimation of shift currents

In this section, we show the numerical estimation of shift charge- and spin-current for the systems discussed in the main text. We restore the fundamental constants (e and \hbar) back to the current expressions, and obtained expected numerical values in experiments.

Shift charge-current in warped Dirac surface states

The warped Dirac Hamiltonian is:

$$H = -v_F \hbar k_y \sigma_x + v_F \hbar k_x \sigma_y + \lambda(k_x^3 - 3k_x k_y^2) \sigma_z \quad (91)$$

$$= -v_F p_y \sigma_x + v_F p_x \sigma_y + \frac{\lambda}{\hbar^3} (p_x^3 - 3p_x p_y^2) \sigma_z \quad (92)$$

where $p_j = \hbar k_j$. We will use $v_F = 2.55(eV \text{ \AA}) = 3.87 \times 10^5(m/s)$ and $\lambda = 250(eV \text{ \AA}^3)$ following the estimation done by Fu [33]. Current operators are:

$$\hat{j}_{para}^i = \frac{\partial H}{\partial A_i} \Big|_{\vec{A}=0} = -e \frac{\partial H}{\partial p_i} = -\frac{e}{\hbar} \frac{\partial H}{\partial k_i} = -\frac{e}{\hbar} v^i \quad (93)$$

$$\hat{j}_{j,dia}^l = \frac{\partial^2 H}{\partial A_l \partial A_j} \Big|_{\vec{A}=0} A^l = e^2 \frac{\partial^2 H}{\partial p_l \partial p_j} A^l = \frac{e^2}{\hbar^2} \frac{\partial^2 H}{\partial k_l \partial k_j} A^l = \frac{e^2}{\hbar^2} (\partial_{k_j} v^l) A^l \quad (94)$$

The susceptibility is:

$$\chi_j^{il} = \frac{1}{\hbar^2 \Omega^2} \int d^2 \vec{p} d(\hbar \omega) Tr[\hat{j}_{para}^i G(\omega + \Omega, \vec{p}) (\hat{j}_{j,dia}^l / A^l) G(\omega, \vec{p})] \quad (95)$$

And, the shift current density is:

$$J_j^{il} = \frac{E^i E^l}{\hbar^2 \Omega^2} \int d^2 \vec{p} d(\hbar \omega) Tr[\hat{j}_{para}^i G(\omega + \Omega, \vec{p}) (\hat{j}_{j,dia}^l / A^l) G(\omega, \vec{p})] \quad (96)$$

As a result, we have the shift current density:

$$J_{j,warp}^{il} = \left(\frac{3e^3 \lambda}{16 \hbar^3 v_F^2} \right) E^i E^l = \left(\frac{3e^3 \lambda}{16 \hbar^3 v_F^2} \right) \frac{2I_0}{\epsilon_0 c} \simeq 0.13 I_0 (nA/m) \quad (97)$$

where I_0 is intensity of light, ϵ_0 is permitivity, c is the speed of light. To make a comparison with experiments, it is convenient to think about current per unit intensity. And, the evaluation of this is dependent of beam size shined on samples. Let us take diameter of the beam size $l = 1(mm)$ which is used in recent experiment by Okada et al. [42] and also in the review article by Ganichev and Prettl [47] (page 956) for the maximally focused beam size. Thus, shift current per intensity for size $1(mm)$ is:

$$\frac{J_{j,warp}^{il}}{I_0} = \frac{J_{j,warp}^{il}}{I_0} = 0.13 (nA W^{-1} \cdot m) \times 1(mm) = 1.3 (nA W^{-1} cm^2) \quad (98)$$

where we take the unit $(AW^{-1} cm^2)$ following Okada et al. [42]. In their experiment, they obtained photocurrent in the order of $10(pA W^{-1} cm^2)$ (page 5 of their arxiv preprint). Our value is two orders of magnitude larger. It is possible that the assumption, photocurrent = (current-density) \times (beam-size), is overestimating actual current measured through leads.

On the other hand, compared to Lindner et al.'s work [48], where they have a photocurrent density $10^{-8}(A/m)$ with sunlight ($I_0 \simeq 140(W/m^2)$ as a modest estimation of sunlight intensity on Earth), our current density value $J_{j,warp}^{il} \simeq 0.13 \times 140(nA/m) = 1.8 \times 10^{-8}(nA/m)$ is in the same order. While they introduced a magnetic strip patterning on TI surface to break both the time-reversal symmetry and in-plane rotational symmetry, we only require warping effect which is naturally present in discovered topological insulators.

Also, our value is also very comparable with the theoretical estimation made by Hosur in circular photo-galvanic effect. His estimation is $100(nA/mm)$ with a $1(W)$ laser for $10(T)$ of in-plane magnetic field. If we assume that the size of sample is $1(mm^2)$, the intensity of light is $I_0 = 1(W)/1(mm^2) = 10^6(W/m^2)$. Then, our photocurrent density is $0.13 I_0 (nA/m) = 0.13 \times 10^6 \times 10^{-3}(nA/mm) = 130(nA/mm)$.

Shift spin-current in gapped Dirac surface states

Shift spin-current is carried in Dirac surface states with band bending and magnetic ordering that breaks the time-reversal symmetry. Hamiltonian is:

$$H = -v_F \hbar k_y \sigma_x + v_F \hbar k_x \sigma_y + m \sigma_z + g \hbar^2 (k_x^2 + k_y^2) \sigma_0 \quad (99)$$

$$= -v_F p_y \sigma_x + v_F p_x \sigma_y + m \sigma_z + g (p_x^2 + p_y^2) \sigma_0 \quad (100)$$

The induced photocurrent density is:

$$J_{j, \hat{s}_n}^{il} = \frac{e^3 g m}{2 v_F (\hbar \Omega)^2} E_i E_l = \frac{e^3 g m}{2 v_F (\hbar \Omega)^2} \frac{2 I_0}{\epsilon_0 c} \simeq 0.31 I_0 (nA/m) \quad (101)$$

where we use $m = 30(meV)$, $\hbar \Omega = 60(meV)$, and band bending parameter $g = 0.36 eV \text{\AA} / (\hbar^2 k_c) \simeq 1.0 \times 10^{30} (kg \cdot m/s)^{-2}$. The band bending parameter is estimated from the ARPES spectrum by King et al. [46] which will be further discussed in the next section. Here, note that the induced shift spin-current is in the same order with the shift charge-current estimated in the previous section for typical choices of Zeeman coupling and photon frequency.

Shift spin-current in 2d electron gas

It is interesting that there are 2d electron gas confined in the surface of 3d topological insulators by a large band bending [45, 46]. King et al. [46] provided numerical values of Rashba coupling strength and effective mass. Consider a Hamiltonian:

$$E^\pm(k) = E_0 + \frac{\hbar^2 k^2}{2m^*} \pm \alpha k \quad (102)$$

In the experiment [46], it is observed that $\alpha = 0.36(eV \text{\AA})$. Effective mass m^* can be estimated by finding $k_c \simeq 0.05(\text{\AA}^{-1})$ in their spectrum such that:

$$\frac{\hbar^2 k_c^2}{2m^*} - \alpha k_c = 0 \quad (103)$$

Thus, the band-bending parameter is:

$$g = \frac{1}{2m^*} = \frac{\alpha}{\hbar^2 k_c}, \quad (104)$$

and, $v_F = \alpha/\hbar$ in terms of parameters in Hamiltonian in the previous section. Again, assuming Zeeman term $m = 30(meV)$ and $\hbar \Omega = 60(meV)$,

$$J_{j, \hat{s}_n}^{il} = \frac{e^3 g m}{2 v_F (\hbar \Omega)^2} \frac{2 I_0}{\epsilon_0 c} = \frac{e^3 m}{2 k_c \hbar^3 \Omega^2} \frac{2 I_0}{\epsilon_0 c} \simeq 1.5 I_0 (nA/m) \quad (105)$$

which is an order of magnitude larger than the shift currents induced by topological surface states.